



Response to Neutrons and γ -rays of Two Liquid Scintillators

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ABSTRACT

UltimaGold™ AB and OptiphaseTrisafe are two liquid scintillators made by Perkin Elmer and EG & G Company respectively. Both are commercially promoted as scintillation detectors for α and β particles. In this work, the responses to γ -rays and neutrons of UltimaGold™ AB and OptiphaseTriSafe liquid scintillators, without and with reflector, have been measured aiming to use these scintillators as γ -rays and neutron detectors. Responses to γ -rays and neutrons were measured as pulse shape spectra in a multichannel analyzer. Scintillators were exposed to gamma rays produced by ^{137}Cs , ^{54}Mn , ^{22}Na and ^{60}Co sources. The response to neutrons was obtained with a $^{241}\text{AmBe}$ neutron source that was measured to 25 and 50 cm from the scintillators. The pulse height spectra due to gamma rays are shifted to larger channels as the photon energy increases and these responses are different from the response due to neutrons. Thus, UltimaGold™ AB and OptiphaseTrisafe can be used to detect γ -rays and neutrons.

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Introduction

Outside the atomic nucleus neutrons are unstable. Neutrons are produced artificially in anthropogenic activities, as well as naturally, mainly during the interaction of cosmic rays with nuclei in the atmosphere; also, neutrons are produced during thunderstorm storm, as well as in nuclear reactions between alpha particles and the nuclei of the earth [1, 2].

In some interactions with matter neutrons generate secondary charged particles and photons. Due to its radiobiological efficiency and the way in which it interacts with matter, it is important to determine, by means of calculations and/or measurements, the neutron energy distribution or neutron spectrum [3]. In order to measure the neutron spectrum it is necessary to use neutron detectors such as activation foils, proportional counter ($^{10}\text{BF}_3$ and ^3He), bubble detectors, inorganic, and organic scintillators [4, 5].

Organic scintillators (liquid or plastic) have fluorescent materials dissolved in a solvent base. Fluorescent materials are aromatic hydrocarbons whose mean atomic number varies from 3.5 to 5. The most commonly used fluorescent materials are the PTP ($\text{C}_{18}\text{H}_{14}$), B-PBD ($\text{C}_{24}\text{H}_{22}\text{N}_{20}$), PPO ($\text{C}_{15}\text{H}_{11}\text{NO}$), and POPOP ($\text{C}_{24}\text{H}_{16}\text{N}_2\text{O}_2$). In the interaction of ionizing radiation with the fluorescent organic materials released energy excites the aromatic solvent molecules. Neutrons are used in basic science (which involves raising and answering research questions), also are used in fusion, fission, to detect special nuclear materials, in dosimetry and characterization of materials [5].

Knowing the fluence rate (flux) and energy distribution of neutrons is important in its use as a neutron source in molten salt nuclear reactors [6]. Also, during calibration and neutron analysis of deuterium plasma experiments it is important to have methods to measure neutrons [7]. In fusion with deuterium reactions, ^{235}U cameras are used to

monitor the neutron flux, spectrometers with Stilbene scintillators to determine the neutron spectrum and to calibrate these detectors, a ^{252}Cf source is used [8]. In certain experiments with fusion, the neutron fluence is usually of the order of 10^{14} n/cm² and it is necessary to look for detectors that support this intensity of radiation [9].

Despite being a mature discipline, neutron detection is always important issue in several areas. Thus, updated measurements of neutrons produced in $\text{U}^{235}(\text{n}, \text{f})$ reactions induced with neutrons from 0.7 to 20 MeV have been reported [10]. Also, ^4He gas scintillators have been used to detect neutrons produced during the nuclear fission of natural uranium samples bombarded with 2.45 MeV neutrons produced in deuterium-deuterium reactions [11].

In problems related with nuclear safety such as: the control and prevention of illicit traffic of Special Nuclear Materials, the characterization of transuranic waste, in safeguards, and in the decontamination and dismantling of nuclear facilities, it is important to measure neutrons. Ports, customs and access points in several countries use gantry detectors with ^3He proportional detectors; however, its scarcity worldwide has led to the need to search for new detection options [12, 13]. Guzmán-García et al, characterized the performance of scintillators of ZnS (Ag) and ^{10}B and determined their response to neutrons produced by sources of $^{241}\text{AmBe}$ and ^{252}Cf [14]. Agreements on the control of nuclear weapons require inspection procedures where neutron detectors like those based on bubble detectors can be used [15].

Personal neutron dosimetry faces several challenges since there is a strong dependence between the angular and energy distribution of the neutrons with the coefficients of neutron fluence-to-dose conversion. The need to overcome these difficulties motivates the search for new solutions, thus in nuclear power plants have been used a naked ^3He thermal neutron detector and two spherical moderators (3 and 9 inches-diameter) to measure thermal, epithermal, and fast neutrons [16]. Around linear accelerators for medical uses silicon diodes type P-I-N have been used to verify the dose due to photoneutrons [17].

Regardless the area where neutrons are used is important to have a reliable method for measuring them and to estimate the dose. The need to measure neutrons has led to innovation in the detection of neutrons, thus Amaro et al, used $^{10}\text{BF}_3$ detectors where ^{10}B was added in aerosol of nanoparticles [18]. Moderate ^3He detectors with high and low density polyethylene are used to measure neutrons produced in the delayed β neutron decay or β -n emission, in order to have evidence that supports certain theories of astrophysics

related to the abundance of the isotopes [19]. Large size liquid scintillators are used in experiments to measure solar neutrinos, proton decay, and dark matter [20]. The DarkSide-50 system has 30 tons of liquid scintillator where background signals, terrestrial and cosmic origin, are suppressed [21].

In the last decade, neutron detectors have been developed to be used in different conditions [22]. The use of liquid scintillators, when they are built in large sizes, have the disadvantage of liquid leaks; there are chemical risks in their handling, and their toxicity, among others; most of these difficulties are eliminated if plastic scintillators are used such as EJ-299-33A [23].

Commercially there are several types of scintillation liquids, such as the UltimaGoldTM AB scintillator liquid and the Optiphase Trisafe scintillator liquid. These are well known in the area of research for the detection of betas and alphas, they are used more routinely, to make measurements of environmental samples. The liquid scintillator UltimaGoldTM AB was used to optimize the determination of ^3H in aqueous samples [24]. Also, this scintillator was used to determine the concentration of Po-210, which is radiotoxic, in tobacco from India [25]. Broda et al, studied the influence of cocktail composition on the standardization of radionuclides [26]. Also, several scintillators were used to determine their response to $^3\text{H}/^{14}\text{C}$ [27]. Another type of commercial liquid scintillator has been used to measure the radon-to-radium ration [28], and in the area of biomaterials [29].

UltimaGoldTM AB and OptiphaseTrisafe scintillator liquids are marketed and promoted for the detection of alpha and beta particles. To be used, the sample must be mixed with the scintillator liquid, therefore both scintillators have low density. If γ -rays interact with these liquid scintillators, electrons will be produced due to photoelectric effect, Compton scattering and pair production, due to the low Z of scintillators the most probable interaction will be Compton scattering, where the scattered electron will induce scintillations. If fast neutrons collide with the scintillators (n, p) reactions will be produced in the hydrogen and the proton will induce scintillations that will have different features than the scintillations induced by electrons. This difference will be noticed in the pulse height spectra, thus both scintillators can be used to detect γ -rays and neutrons. Therefore, the objective of this work was to determine the response to gamma rays and neutrons of two organic liquid scintillators that are commercialized as detectors for beta and alpha particles.

Materials and Methods

In this study, we used two scintillation liquids commercially promoted as alphas and betas detectors. These detectors are UltimaGold™ AB and OptiphaseTriSafe, the first is from PerkinElmer and the second is from the company EG & G company.

Both scintillation liquids are organic compounds whose content of carbon and hydrogen is high, therefore should detect gamma rays by the excitation produced by the electrons released in the scintillator and detect neutrons by the excitation produced by the protons released by fast neutrons in (n, p) reactions

with hydrogen in the same way that alphas induce scintillation.

In order to determine the response of both scintillators to gamma and neutron, two 25 ml containers were prepared, one with each scintillator liquid. The containers are made of glass with low potassium content, since potassium absorbs the type of light that is emitted by the scintillator liquid. The liquid scintillator container was optically coupled to a photomultiplier tube (PMT) Dupont brand model 6292, as is shown in Figure 1 (a).



(a)



(b)

Figure 1: (a) Vial with the scintillator coupled to the PMT.
 (b) Measuring the response to ^{60}Co gamma-rays.

The PMT has a base made by EG & G Ortec model 296. This array was connected into a spectrometric system with a high voltage supply, an amplifier, and a multichannel analyzer. The scintillator container and the PMT were isolated from the light using a thick black polyethylene bag and the response to gamma rays was carried out allocating the γ -ray source above the scintillator container, as is shown in Figure 1 (b).

The pulse height spectra of 0.511, 0.662, 0.834, 1.17, 1.27, and 1.33 MeV γ -rays produced by ^{137}Cs , ^{54}Mn , ^{22}Na , and ^{60}Co sources was measured by both scintillators using the same measuring conditions. Gamma-ray sources were produced by Eckert & Ziegler Isotope Products. On November 15th, 2015 the initial activity of the sources are shown in Table 1.

Table 1: Gamma ray sources initial activity

| Sources | Initial activity [kBq] |
|-------------------|---------------------------|
| ^{137}Cs | $38.92 \pm 3\%$ |
| ^{54}Mn | $40.03 \pm 3\%$ |
| ^{22}Na | $35.67 \pm 3\%$ |
| ^{60}Co | $38.79 \pm 3\%$ |

For each of these sources, and for radiation background, the pulse height spectrum was measured in the multichannel analyzer.

In order to determine the response to neutrons, a source of $^{241}\text{AmBe}$ of $3.7 \times 10^9 \pm 10\%$ Bq was used. The pulse height spectrum due to neutrons was measured to 25 cm and 50 cm source-to-scintillator distances.

Background, gamma ray, and neutron pulse height spectra were measured using 1024 channels and during a live time of 1800 seconds, using the same electronics features conditions.

The first group of measurements was carried out with

no reflector in the scintillator liquid containers. Then, glass containers were painted using nail polish in white, and background, γ -rays and neutrons pulse height spectra were measured again.

Results

In Figure 2 are shown the responses of UltimaGold™ AB scintillator without reflector. The response is shown in terms of the pulse height spectra due radiation background, ^{137}Cs , ^{54}Mn , ^{22}Na , and ^{60}Co -rays, and to neutrons produced by the $^{241}\text{AmBe}$ isotopic neutron source. The response to neutrons is to 25 and 50 cm from the scintillator.

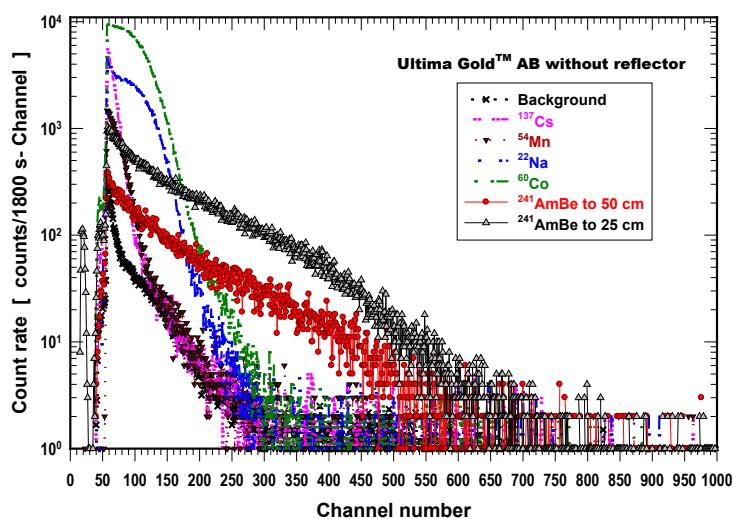


Figure 2: Pulse height spectra, due to background, γ -rays, and neutrons, measured with Ultima Gold AB scintillator without reflector.

In Figure 3 are the pulse height spectra produced by the scintillator OptiphaseTrisafe without reflector, while in Figure 4 the responses of UltimaGold™ AB

scintillator with reflector. Responses are the pulse height spectra due to background, γ -rays, and neutrons.

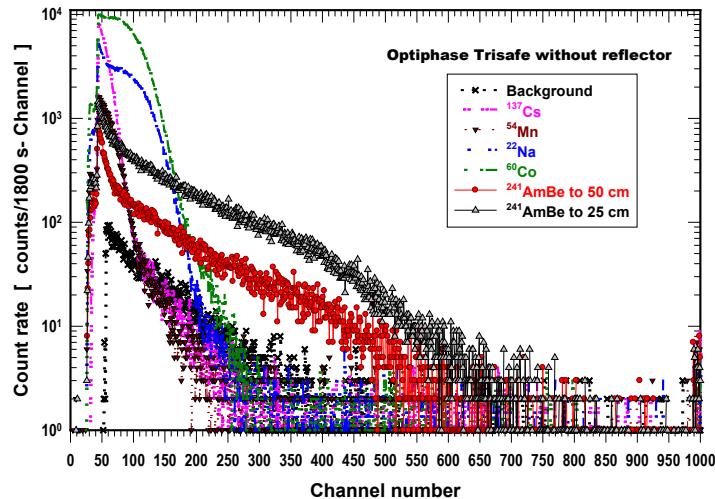


Figure 3: Pulse height spectra, due to background, γ -rays, and neutrons, measured with OptiphaseTrisafescintillator without reflector.

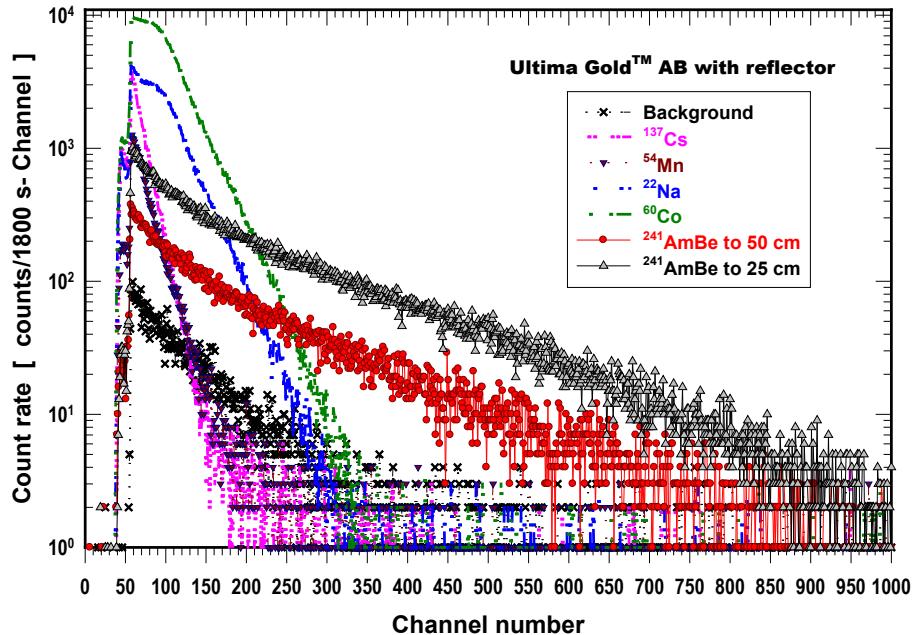


Figure 4: Pulse height spectra, due to background, γ -rays, and neutrons, measured with Ultima Gold AB scintillator with reflector.

In Figure 5 are the pulse height spectra produced by the scintillator OptiphaseTrisafe with reflector.

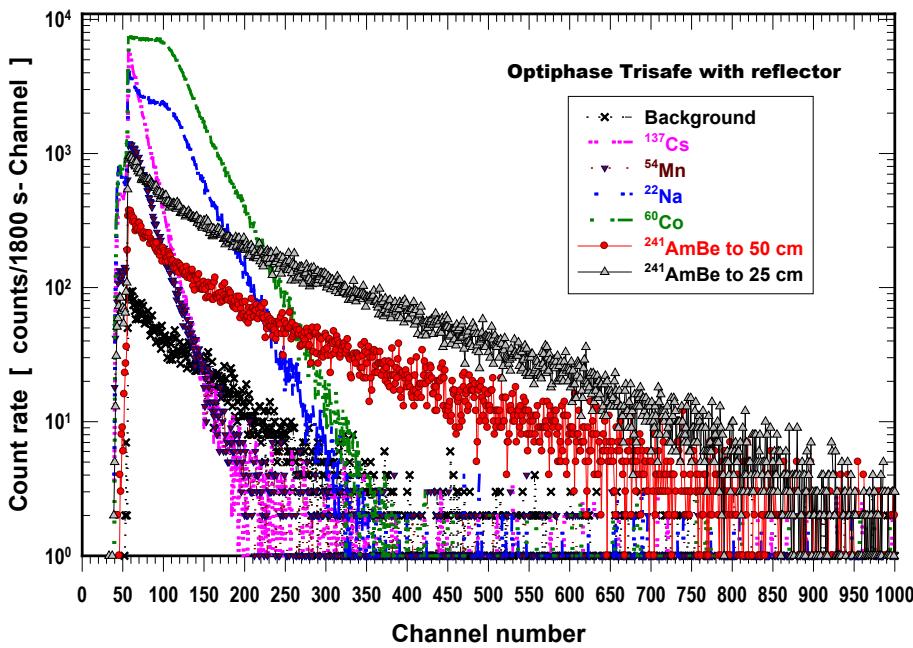


Figure 5: Pulse height spectra, due to background, γ -rays, and neutrons, measured with OptiphaseTrisafe scintillator with reflector.

In figure 6 are shown the pulse height spectra (PHS) of ^{137}Cs and ^{60}Co γ -rays on UltimaGoldTM AB scintillator without and with reflector. For the OptiphaseTrisafe scintillator, without and with reflector, in figure 7 are shown the spectra of ^{137}Cs and ^{60}Co γ -rays. In Figures

6 and 7 the PHS have been corrected by background. The effect, on the PHS due to neutrons, of adding the reflector in both scintillators is shown in Figure 8 for UltimaGoldTM AB and in Figure 9 for Optiphase-Trisafe.

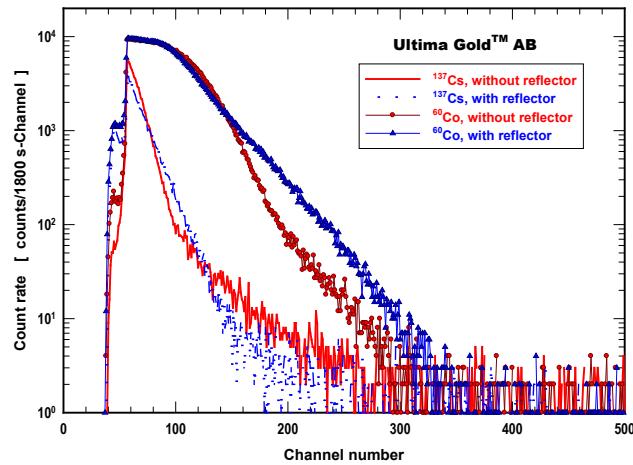


Figure 6: Background corrected pulse height spectra, due to ^{137}Cs and ^{60}Co γ -rays, measured with Ultima-Gold™ AB, without and with reflector.

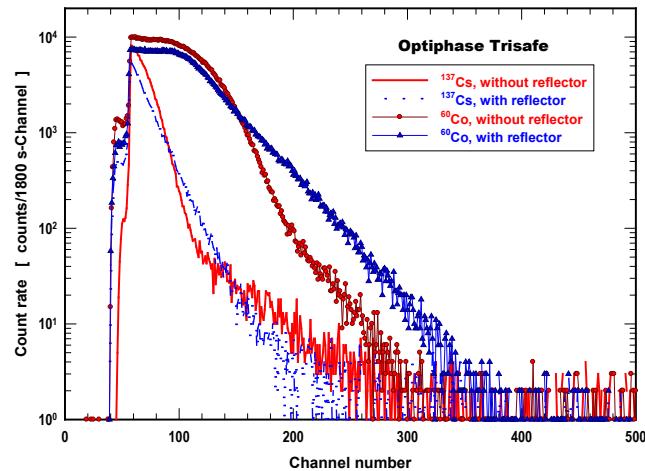


Figure 7: Background corrected pulse height spectra, due to ^{137}Cs and ^{60}Co γ -rays, measured with Optiphase-Trisafe, without and with reflector.

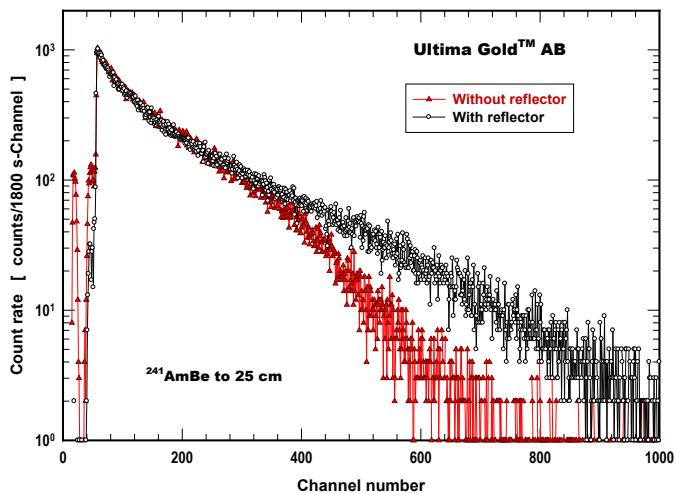


Figure 8: Background corrected PHS due to $^{241}\text{AmBe}$ neutrons measured with UltimaGold™ AB scintillator without and with reflector.

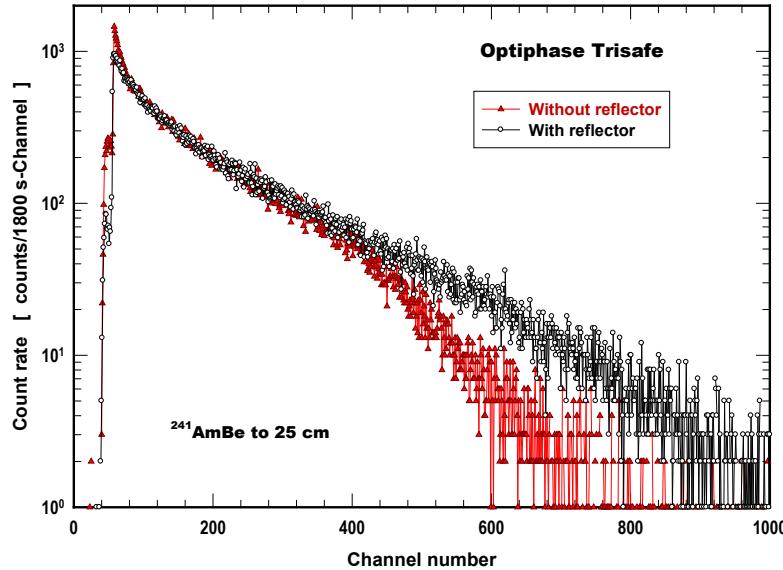


Figure 9: Background corrected PHS due to $^{241}\text{AmBe}$ neutrons measured with OptiphaseTrisafe scintillator without and with reflector.

Discussion

In the Figures 2 and 3 show the pulse height spectra for both scintillators, without reflector, for the background, gamma rays and neutrons. The pulse height spectra for neutrons are different to the pulse height spectra for γ -rays. In the responses to gamma-rays the Compton edge shifts to the right as the gamma ray energy increases. For γ -rays of ^{60}Co practically the pulse height spectrum ends in channel 350.

In the case of neutron responses, for both scintillators the pulse height spectra have the same shape from channel 50 to channel 750. However, the pulse height spectrum is greater when the source is to 25 cm than when it is to 50 cm. This is due to the neutron flux at 25 cm is greater than the neutron flux at 50 cm distance. The response of the scintillators for gammas is different than the response for neutrons. This is because the interaction of the gamma rays is mainly with the electrons of the scintillators, while the interaction with neutrons occurs with the hydrogen of both scintillators.

In Figures 4 and 5 the pulse height spectra for background, γ -rays and neutrons for both scintillators with reflector are shown. Here can be noticed that the shape and end point of pulse height spectra for gamma-rays are the same as the case of scintillators without reflectors. The shape of pulse height spectra for neutrons looks alike the spectra measured without reflector, however, when the reflector is used the end point is in channel 1000. Therefore, the use of reflector increases the amount of scintillations reaching the PMT.

The shape of pulse height spectra (PHS) for γ -rays and neutrons (Figures 2, 3, 4, and 5) are consistent with those reported by Wang, Seidaliev & Mandapaka for their design of a neutron rem meter [30].

The PHS for $^{241}\text{AmBe}$ neutrons of both scintillators are similar to the pulse height spectra reported by Beccetti et al, although they used C6D6 scintillators; NE230, BC537 and EJ315 which are plastic scintillators, and an organic scintillator in liquid state of deuterated Xylene (C8D10; EJ301D) [31].

The responses of both scintillators to neutrons are similar to those reported for a plastic scintillator EJ-299-33A that together with BC501A were irradiated with almost monoenergetic neutrons [23].

For both scintillators, without or with reflector, as the γ -ray energy increases the pulse height spectrum shifts to higher channels. For 0.662 MeV γ -rays can be noticed that approximately from channel 80 to channel 120 the pulse height spectrum is larger when measured with the UltimaGoldTM with reflector. The same effect is shown for ^{60}Co γ -rays where the increase is noticed approximately from channel 150 to channel 320 (Fig. 6). For Optiphase (Fig. 7) the same effect is noticed with the difference that for ^{137}Cs photons a larger amount of counts are notice from channel 90 to 140 approximately and from 160 to 340 for ^{60}Co γ -rays.

The effect of adding the reflector is also noticed, for both scintillators, in the PHS corrected by background for the $^{241}\text{AmBe}$ neutrons. For both scintillators the spectrum measured with the reflector has a larger amount of counts per channel above channel 400. In the case of UltimaGoldTM beyond channel 400 the count

rates measured with the scintillator with reflector are larger than those obtained without the reflector (Fig.8). This difference is larger than the Optiphase Trisafe scintillator (Fig. 9).

Conclusions

In this work have been measured the gamma and neutron responses of the two organic scintillation liquids, UltimaGold™ AB and OptiphaseTrisafe, which are promoted commercially to measure β and α particles.

The two organic liquid scintillators, UltimaGold™ AB and Optiphase Trisafe, in addition to be used to measure α and β particles, can be used to measure γ -rays and neutrons, because the PHS due to γ -rays is different to the PHS due to neutrons.

The PHS due to gamma rays show the Compton edge that shifts to the right as the energy of the incident photon increases. As the photon energy increases the PHS end-point shifts to the right. The end-point for ^{60}Co γ -rays is around channel 350.

The PHS due to neutrons has different shape to the PHS due to γ -rays. For neutrons the PHS end-point is in channel 1000.

For both scintillators the use of a white color reflector improves the amount of count rates in the PHS.

A limitation of this work was that to measure neutrons a $^{241}\text{AmBe}$ neutron source was used. Beside neutrons this source produce 4.4 γ -rays due to $^{12}\text{C}^*$ decay that is produced during the $\text{Be}(\alpha, n)^{12}\text{C}^*$ reaction, and 59.5 keV γ -rays during the ^{241}Am decay. Also the neutron source is enclosed in a shell of polyethylene where neutrons are moderated and captured by H producing 2.2 MeV photons. Therefore, in the neutron pulse height spectra count rates are due to gammas and neutrons. If the pulse shape discrimination technique is not available, the scintillators can be used to measure neutrons by assuming that beyond channel 400 the count rate are only due to neutrons.

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